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A function using cubic splines for the analysis of alpha-particle spectra from silicon detectors

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Abstract

A function based on the characteristics of the alpha-particle lines obtained with silicon semiconductor detectors and modified by using cubic splines is proposed to parametrize the shape of the peaks. A reduction in the number of parameters initially considered in other proposals was carried out in order to improve the stability of the optimization process. It was imposed by the boundary conditions for the cubic splines term. This function was then able to describe peaks with highly anomalous shapes with respect to those expected from this type of detector. Some criteria were implemented to correctly determine the area of the peaks and their errors. Comparisons with other well-established functions revealed excellent agreement in the final values obtained from both fits. Detailed studies on reliability of the fitting results were carried out and the application of the function is proposed. Although the aim was to correct anomalies in peak shapes, the peaks showing the expected shapes were also well fitted. Accordingly, the validity of the proposal is quite general in the analysis of alpha-particle spectrometry with semiconductor detectors.

1. Introduction

The analysis of spectra obtained from silicon semiconductor detectors is a crucial task in alpha-particle spectrometry. When the peaks in these spectra are well separated, the determination of important parameters such as areas and peak positions is readily accomplished. However, a peak in an alpha spectrum from this type of detector has asymmetric width, with tails extending from the maximum to the low-energy side [1]. These tails may become quite long, such that overlapping often occurs between peaks.

This effect is more severe when the peaks in the spectra belong to emissions with very similar energies and the absorption undergone by alpha particles is more important: thick sources (self-absorption at the source), air pressure in the counting camera, etc. [2].

Over the years, several analytical functions have been proposed to parametrize this asymmetric shape of the peaks, obtaining very good results in the fitting and deconvolution of overlapping peaks. Many of these functions are able to reproduce very high degrees of asymmetry through the use of several asymmetric terms in their definitions. However, in practically all cases such definitions assume

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that the peaks have the natural shape expected of those obtained with silicon detectors, which has been determined using suitably obtained sources and very well controlled conditions in the measurements.

Commonly, the peaks in alpha spectra obtained from natural samples are also well fitted by these analytical functions, but sometimes changes in the counting conditions induced by very long counting times or very poor characteristics in the source deposits do not allow such analytical functions to properly fit the peak shapes observed, causing important errors in the evaluation of the activity of the radionuclides present in the source [3].

In such cases, the origin of function insufficiency is mainly due to the difficulty in alpha-particle spectrometry to foresee the existence of anomalies and their relative contributions to the final peak shape.

Therefore, a function with high flexibility but without an a priori defined shape could be desirable to analyze such irregularities. Cubic splines curves are able to cope with these requirements since they are based on the distribution of several control points (knots) which may or may not belong to the data distribution to be fitted [4]. Here, these properties were used to construct a function for alpha-particle spectral analysis, although certain important modifications were introduced in the common definition of the curves to make them amenable to the task in hand: alpha spectra are frequently the sum of several overlapped peaks and hence the knots of the splines curve do not necessarily belong to the experimental distribution of data.

On the other hand, these curves are too flexible and the results may be flawed if no constraints are imposed in their application to alpha peak fitting.

The above anomalies render the peak shapes different from those expected from silicon detectors although the gross characteristics of the peaks are preserved, mainly at the edges of the peaks: left-hand side tails; almost Gaussian shapes on the right-hand side of the maximum, etc. The usual anomalies appear as bumps on the left-hand side and close to the peak maximum, caused by slight gain drifts, pressure changes during counting, fluctuations in the experimental data, etc. [5].

In this paper we present a function based on these characteristics of the peaks that has been modified by the introduction of a cubic splines term in the intermediate region. The function allows us to reproduce the irregularities sometimes observed in the peak shapes.

Some ideas about the application of this type of very flexible curves to the analysis of alpha-particle spectra have already been proposed by Martin [3]. With regard to these basic ideas, detailed study of the function proposed by this author suggested some important modifications that might improve the application of this type of function to the analysis of alpha-particle spectra obtained with silicon semiconductor detectors.

These modifications mainly involve certain initially considered parameters which caused severe instabilities in the minimization process and also strongly hindered the initialization required for the optimization of a function with non-linear dependence on its parameters.

Other considerations related to the minimization algorithm led us to introduce SVD as a powerful method to gain more stability and at the same time to reduce the importance of the initialization. Moreover, it ensures convergence in the process.

In Section 2, the function is described in detail. Certain other aspects regarding the program constructed to apply this function are dealt with in Section 3.

In Section 4, the results of tests carried out to determine the reliability of the program using the proposed function are shown. A detailed discussion of these results is also offered.

Section 5 presents the most important conclusions.

2. Descriptive remarks

As has been mentioned, it is well known that the peaks in alpha-particle spectra from silicon semiconductor detectors have an asymmetric shape with more or less long tails towards the low-energy side. Several authors have studied these shapes in order to reproduce the alpha lines using analytical functions based on semi-theoretical considerations. Thus, the basic response of the detector to the arrival of an alpha particle is essentially Gaussian.

This basic shape is also obtained when a beam of monoenergetic alpha-particles interacts with the detector: energy straggling, electronic noise of the chain, etc. [6].

It has repeatedly been demonstrated that the peaks in spectra obtained from these detectors can be described by a linear combination of increasing left-hand side exponentials truncated at the energy of the alpha particles. These exponentials represent all the phenomena producing absorption of the particle energy in the path from the emission site to the sensitive detector (self-absorption at the source, absorption in the dead layer of the detector window, absorption in the counting camera due to scattering with air particles, etc).

Bortels and Collaers [7] proposed a function based on the above considerations, in which the final terms are constructed by the convolution of the basic response with the linear combination of truncated exponentials. The number of exponential terms depends on the requirements of the experimental peaks, but two are usually enough to reproduce most experimentally obtained spectra.

A similar proposal has been made by Wätzig and Westmeier [8], but in this case the authors included an additional Gaussian term in the description of the function: this is obtained from the convolution of the basic response of the system (Gaussian) with a delta distribution (ideal response for monoenergetic alpha particles). Other authors have also found that at the left of the maximum the junction of several polynomial forms with Gaussians produce good results [9]. Accordingly, in nearly all cases it is well accepted that the alpha peaks from silicon semiconductor detectors present a Gaussian form at the right-hand side of the maximum. At the left and far from the maximum, the shape can be represented by an exponential. These characteristics are very useful because they serve to provide the end conditions to the cubic splines term introduced in the function proposed here, as will be described below.

In a log-normal representation of the alpha peaks, it is easily seen that at the edges the peak can be fitted by a positive-slope straight line and a parabolic curve to the left-hand and right-hand sides of the maximum, respectively, corresponding to the increasing exponential and the Gaussian shape on a normal–normal scale (Fig. 1). Between these two regions, the peak shows an increasing slope with a degree of curvature that strongly depends on the absorption effects undergone by the alpha particles. In any case, the peak in the complete interval shows a smooth aspect, which in semilogarithmic representation is softer than on a linear scale.

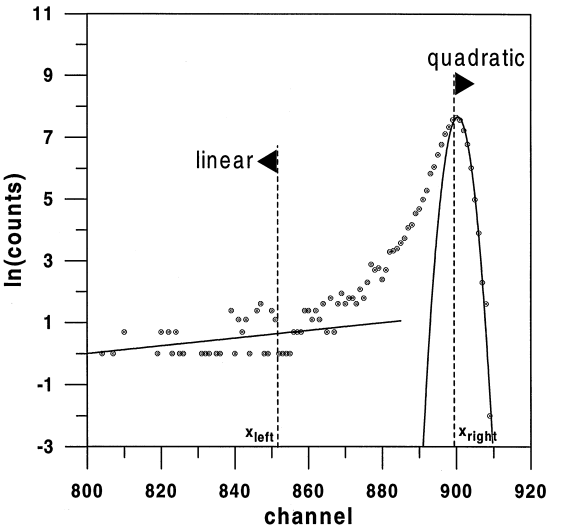

Fig. 1. Semilogarithmic representation of an alpha-particle spectrum with one peak obtained from a silicon surface barrier detector.

In what follows, the function proposed in this work is described assuming that only one peak forms the alpha spectrum. Later, this function will be extended to a more general spectrum formed by several peaks.

If the alpha spectrum consists of one peak produced by an emission of energy E, the semilogarithmic representation of the data distribution ($y(x_i)$) can be described as

$$\ln(y(x_i)) = S(E, x_i).$$

At the edges of the peak, $S(E, x_i)$ describes a positive-slope straight line at the left and a parabolic curve at the right of the maximum, taking into account the considerations already discussed

$$S_L(E, x_i) = A + Bx_i, \quad (x_i < x_{lef})$$

$$S_R(E, x_i) = Q + Px_i + Rx_i^2, \quad (x_i > x_{rig}).$$

Between these two regions, a cubic splines curve is used to attach the linear and quadratic parts. If X_j represents the knots, in the splines region ($X_1 \le x_i \le X_n$, with $X_1 = x_{\text{left}}$ and $X_n = x_{\text{right}}$ as in Fig. 1) the function (using the second derivative formalism) can be written as

$$S_s(E, x_i) = \left(\frac{X_{j+1} - x_i}{X_{j+1} - X_j}\right) Y_j + \left(\frac{x_i - X_j}{X_{j+1} - X_j}\right) Y_{j+1} + \left\{\left(\frac{X_{j+1} - x_i}{X_{j+1} - X_j}\right)^3 - \left(\frac{X_{j+1} - x_i}{X_{j+1} - X_j}\right)\right\} \times \frac{(X_{j+1} - X_j)^2 Y''_j}{6} + \left\{\left(\frac{x_i - X_j}{X_{j+1} - X_j}\right)^3 - \left(\frac{x_i - X_j}{X_{j+1} - X_j}\right)\right\} \frac{(X_{j+1} - X_j)^2 Y''_{j+1}}{6} \quad (1)$$

where Y_j are the ordinates on a logarithmic scale and Y''_j are the second derivatives at the knots X_j.

The knots are defined as

$$X_j = E + r_j \quad (2)$$

where r_j are real numbers giving the relative position of the knots with respect to the energy of the peak. The value of r_j is decided by the user to define the knots, depending on the experimental distribution characteristics.

Taking into account the boundary conditions at the junctions of the cubic spline term with linear and quadratic terms (continuity of the first and second derivatives at these junctions is imposed to reach smoothness), the linear region ($x_i < X_1$) can be described as

$$S_L(E, x_i) = (Y_1 - Y'_1 X_1) + Y'_1 x_i \quad (3)$$

and the quadratic region ($X_n < x_i$) as

$$S_R(E, x_i) = -\frac{1}{2\sigma^2} x_i^2 + \left(Y'_n + \frac{X_n}{\sigma^2}\right) x_i + \left(Y_n - \frac{X_n^2}{2\sigma^2} - Y'_n X_n\right) \quad (4)$$

where σ is the width of the Gaussian (on a linear scale).

Thus, peak distribution in the complete interval is totally described. Accordingly, the fittable shape parameters, common for all peaks in the spectrum if there is more than one, will be the knot heights on the logarithmic scale $\{Y_1 .. Y_n\}$ and the width of the Gaussian σ.

If a spectrum contains NPI peaks, data distribution can be described as the contribution of all peaks in each channel:

$$y(x_i) = \sum_{k=1}^{\text{NPI}} F_k(x_i)$$

where $y(x_i)$ is the content of channel x_i and $F_k(x_i)$ is the contribution of peak k to the same channel on the linear scale.

Taking into account that peak shape is imposed to be the same for all peaks in the interval analyzed, the values of the fittable parameters allow us to construct a model peak with this shape; this model peak is placed in the different positions (E_k) and amplified by a multiplicative factor (H_k), both characteristic of each peak in the spectrum. The E_k and H_k of each peak are also parameters of the fit.

The data distribution of the spectra can be described on the linear scale as:

$$y(x_i) = \sum_{k=1}^{\text{NPI}} H_k e^{S(E_k, x_i)}$$

Determination of the second derivatives in the knots (Eq. (1)) can be accomplished easily by applying an algorithm such as LU decomposition [10]. The linear equation system used to obtain these second derivatives is tridiagonal and hence its resolution is easily computerized.

The first derivatives at X_1 and X_n necessary to compute linear and quadratic intervals (Eqs. (3) and (4)) are calculated as:

$$Y'_1 = \frac{Y_2 - Y_1}{X_2 - X_1} - \frac{X_2 - X_1}{6}(2Y''_1 + Y''_2)$$

$$Y'_n = \frac{Y_n - Y_{n-1}}{X_n - X_{n-1}} - \frac{X_n - X_{n-1}}{6}(Y''_{n-1} + 2Y''_n) \quad (6)$$

In Eq. (6) it can be seen that Y'_1 and Y'_n are established from the boundary conditions at the edges of the splines region. It is therefore not necessary to consider the slope in the right-hand-most knot as a fitting parameter. More about this will be dealt with in Section 4.

3. Minimization

Since the complete function has non-linear dependence on the fittable parameters, an iteractive algorithm starting from an initial set of parameters is necessary. In the context of minimum squared deviations, the Levenberg–Marquardt algorithm is recognized to be one of the most powerful methods to carry out non-linear optimizations [10]. Here, this algorithm was used to minimize the χ^2 figure, which is defined as

$$\chi^2 = \sum_{i=1}^{N} w_i(y_i - F_i)^2$$

where y_i are the counts at channel x_i, F_i are the optimized function values in same channel and w_i are the weightings associated with each channel, which are defined as

$$w_i = \frac{1}{\sigma_i^2}$$

σ_i^2 being the variance from the fitted distribution at x_i. Taking into account several considerations [11] these variances can be calculated as

$$\sigma_i^2 = F_i + 1.$$

The SVD method was implemented to carry out the matrix inversion in the minimization process, thus avoiding potential ill-conditioned matrices or singular situations in the linear equation systems in order to obtain solutions either for each iteration or for the minimum to evaluate the covariance matrix. This method helps to gain stability in the optimization process and reduces the importance of the initialization of parameters in a non-linear minimization. Some remarks about these considerations are given in Ref. [12].

4. Discussion

4.1. Fitting parameters

One of the advantages of the function proposed here is that it does not have a predefined shape, such that it is able to represent an almost arbitrary data distribution, only constrained at the edges of the peaks. In alpha-particle spectrometry, the low reproducibility from counting to counting, due to changes in the operating conditions (different thickness or active areas in the sources, pressure changes in the counting camera, etc.), prevents one from carrying out valid calibrations of the peak shapes. This is a serious problem when analytical functions with many terms are used to fit the spectra because it is very difficult to assign correct initial values to the parameters, sometimes leading to extremely long processes or even no convergence in the fittings. This problem is exacerbated when the proposed functions are overparametrized.

In our program, the definition of the function facilitates the initialization of the parameters. Some of them, such as E_k and H_k, can be evaluated directly from visualization of the spectra assuming approximate energy calibrations and approximate areas for the peaks. On the other hand, the initialization of the shape parameters demands that one peak of the spectra be taken as a reference (Fig. 2), for which the parameter H_{ref} is initialized at 1. The evaluation of σ is very easily done, taking the channel at the right of the maximum where the content is about $Y_{\max}/2$. Estimation of the knot

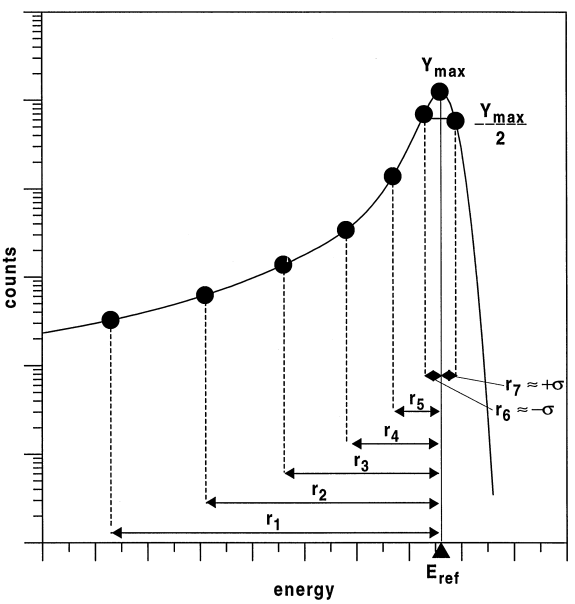

Fig. 2. Estimation of initial parameter values using a peak from the alpha-particle spectrum as reference.

heights on the logarithmic scale is done following approximately the shape of the reference peak (even when this reference peak is overlapped, it is not very difficult to estimate a well-approximated shape). A good degree of approximation in the initial assignment of the parameters is achieved and this considerably improves the optimization process.

According to the program description given in Sections 2 and 3, it is appropriate to discuss certain aspects related to the definition of the function and the minimization process.

The width of the Gaussian (σ) and the boundary conditions at the knot farthest to the right (X_n) completely determine the Gaussian characteristics. This σ, which is a fitting parameter, is necessary to determine the end conditions of the spline function at the right edge and hence the second derivative at the right-hand-most knot. At the left-hand side of the maximum, at X_1, the spline connects smoothly with the straight line, such that the required second derivative at this knot is 0.

Once the second derivatives at all knots are known, the first derivatives at each point of the spline region are readily determined (Eq. (6)). This is very important because it allows one to eliminate the first derivative at the right-hand-most knot as a free parameter, as recommended by Martin [3]. This author located the center of the Gaussian at the energy (E) that characterizes the peak. This constraint reduces the flexibility of the function and requires the introduction of an additional parameter: the slope of the curve at the knot farthest to the right.

In our program, tests taking this slope as a fitting parameter produced strong numerical instabilities. It was very difficult to correctly initialize this parameter and the process proved to be very sensitive with regard to this initial value, in some cases producing very poor solutions.

In our function, either the center of the Gaussian or the slope at X_n is wholly established from the boundary conditions and the width of the Gaussian (σ). This implies an immediate reduction in the number of parameters but leads to more direct optimization processes. Moreover, the initialization of the parameters is less critical and the minimization can be achieved much faster.

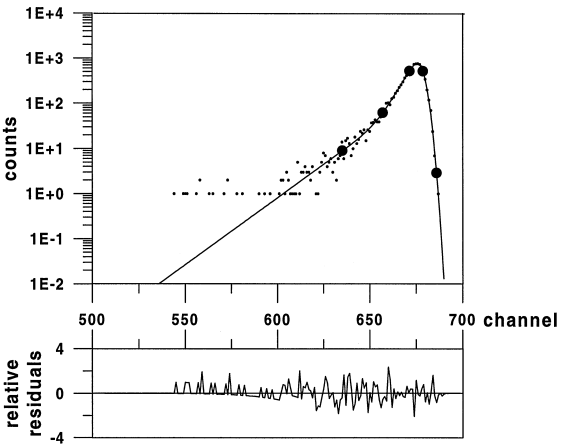

Fig. 3. Fitting of a ^{210}Po peak using only 5 knots.

Visualization of the spectrum also helps to decide about the position of the knots. Martin [3] recommends that they should be taken at positions that are multiples of σ regarding the maximum of the peak, increasing the distance between them for the farthest knots. The vector of real numbers [r] defines the relative location of the knots and these real numbers are fixed values at the beginning of the program.

In this situation, we found that one knot at the right-hand side of the maximum, one close to the maximum, and three more at the left-hand side afforded very successful fits in most spectra with well-conformed peaks (Fig. 3), although the most suitable number of knots strongly depended on peak widths and tails (anomalous peaks or very long tails may require a larger number of knots, Fig. 4). Presently, we have not found a clear rule to define an a priori optimum number of knots because of this dependence but, in general, the necessary number of knots is lower than that found suitable in Ref. [3]. The fitting process rarely requires more than 8 knots, which considerably reduces the total number of parameter in the fitting and leads to quicker optimization processes. One could speculate that the absence of the slope at X_n as a free parameter may be the cause of the lower number of knots required when using the function proposed here.

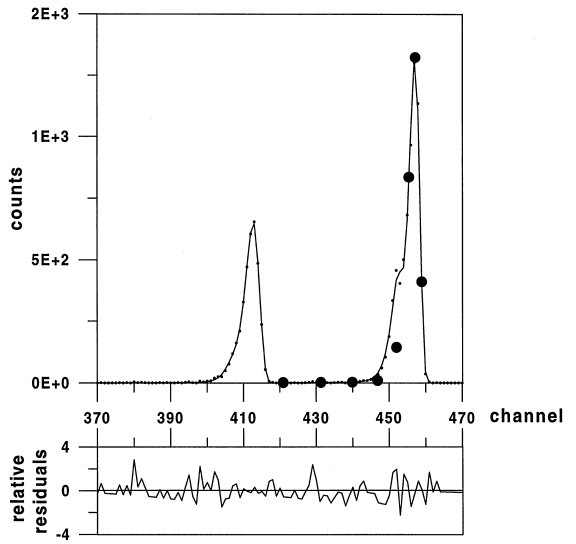

Fig. 4. Alpha spectrum of ^{239}Pu, ^{240}Pu and ^{238}Pu (FWHM was about 27 keV). 8 knots were used to obtain a sufficiently good fit of the data. The most energetic peak of ^{238}Pu was used as reference.

4.2. Optimization process

Routines involved in a program based on the use of cubic splines to fit alpha-particle spectra can be more complicated than those required by other normalized analytical functions [7,13]. An iteration method for optimization, such as the Levenberg–Marquardt method, requires computation of the function and its first derivative with respect to the free parameters at each point of the interval considered [10]. To compute the first derivative of the function, it is first necessary to determine the value of the second derivatives at the knots. From a computational point of view, the problem derives from the fact that those second derivatives depend on some of the fitting parameters.

The problem is easily solved using Richardson's method [14], although this introduces additional calculations into the program. Tests were carried out to confirm the reliability of the implementation of this approximate method on the results. For this, the results obtained by applying Richardson's method were compared to a complete analytical development of the process using the Mathematica ver. 2.0 program. Taking the minimum obtained from the analytical method as a reference, in all cases assayed the precision achieved in this comparison was good enough to consider the solutions obtained with both methods as identical. The worst divergence obtained using 4 knots was 10^{-4} % with regard to the analytical result.

4.3. Determination of peak areas

Peak area determination is another aspect that requires explanation. Usually, this type of program is constructed to determine the activity of radionuclides present in the source, the isotopic composition of the deposit, or the energy of the emissions of such radionuclides. With this function, which is not currently normalized, the area is not a fitting parameter. Therefore, this must be calculated once the spectrum has been fitted.

The sum of the content channel by channel in a selected interval of the spectrum is elementary, but correct determination of the area is not so easy for two reasons: first, it must be ensured that the total area of the peaks is estimated with no significant bias; second, the error associated with the areas requires the computation of all covariances in all channels of the interval. Regarding the first point, choice of the interval to compute the area can be done by visualization, taking the edges of this interval such that the reference peak is guaranteed to be wholly contained in it. This same interval size is considered in the evaluation of the area for the rest of the peaks. Fig. 5 shows this criterion as applied to a spectrum of natural uranium spiked with ^{232}U. Only three peaks are used in this figure to illustrate the criterion. Selection of the interval size for computing the areas is made on the reference peak (the most energetic emission of ^{232}U) and this same interval is considered for the other 23 peaks in the spectrum. Fig. 5 shows the effect of applying this criterion to the left-hand-most visible peak of ^{232}U.

Table 1 compares the results of fitting this spectrum with the function proposed here and a normalized analytical function with two tail terms [15].

Regarding the use of the splines-based function, the results in Table 1 show the effects of truncation at channel 0 and extrapolation of the fit to avoid

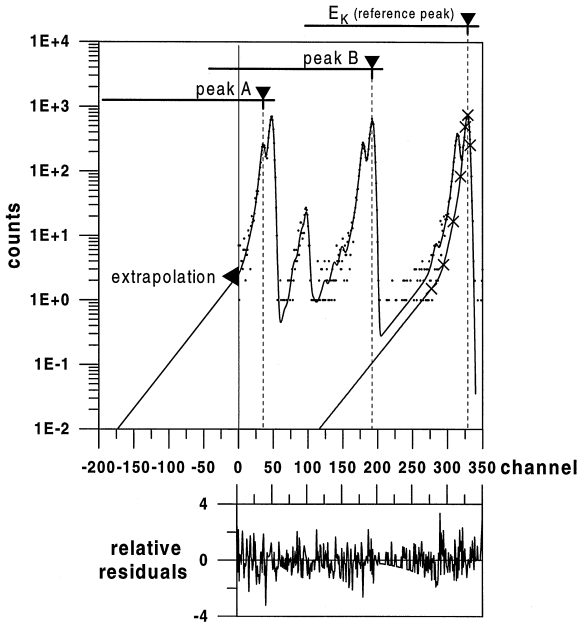

Fig. 5. Alpha spectrum of natural uranium spiked with ^{232}U in which the criterion for area evaluation is shown. The most energetic peak of ^{232}U was used as reference.

significant bias in the calculation of the areas. The residuals obtained using the splines-based function and the normalized function were very similar and only slight differences in the areas were found. χ^2_{red} was also similar for both fittings (close to 0.95).

Truncation at the real limit to the left of the spectrum (channel 0) produced a bias in the total area of ^{238}U, which was conveniently corrected by applying the extrapolation.

The agreement between the experimental total area of the interval and those obtained from the fitting functions in the same range was very good, as can be seen in Table 1.

Determination of the errors associated with the areas was carried out by the application of the error transmission law, although this involved additional calculus that considerably slowed down the overall fitting process. Therefore, the initial choice of the interval for computing the areas must be large enough to avoid significant bias but at same time as small as possible to avoid overly long processes in the determination of their errors. This does not present serious difficulties.

The errors of the areas in the example shown in Table 1 were very similar for the normalized function (the areas were obtained directly from the fit and their errors from the covariance matrix) and for the function proposed here.

4.4. Application to anomalous peaks

The flexibility of the function studied here is clearly seen in Fig. 6, in which the three most important emissions of the ^{232}U nuclide are well distinguished. The fit shown as a solid line was obtained assuming a single peak of energy 5320.3

Table 1
Results from fits using the function proposed in this work and a normalized analytical function on the spectrum of Fig. 5

Nuclide	Calculated area (counts)			Experim. area (counts)
	Our function		Normalized function	
	Truncated	Extrapolated		
^{238}U	7807 ± 89	7880 ± 90	7854 ± 90	
^{235}U	357 ± 23	358 ± 23	360 ± 23	
^{234}U	7485 ± 87	7486 ± 87	7462 ± 87	
^{232}U (rf[a])	9708 ± 112	9708 ± 112	9682 ± 110	
Σ Nuclides	25357 ± 169	25432 ± 170	25358 ± 168	
Region channels: 0–350	25357 ± 170		25304 ± 168	25342 ± 159

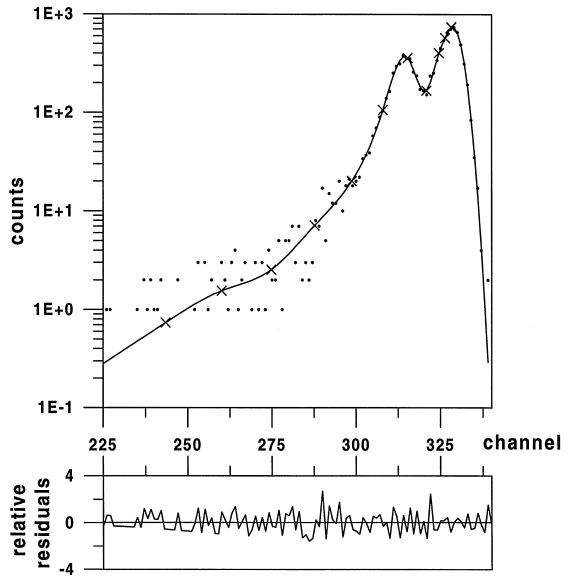

Fig. 6. The ^{232}U region of the spectrum of Fig. 5 was fitted. Only one peak was considered, grouping the three emissions of ^{232}U.

keV instead of the sum of three peaks corresponding to emissions of 5139.0 keV (0.28%), 5263.5 keV (31.2%) and 5320.3 keV (68.6%). In this way, this experimental distribution of the data reveals many changes in curvature between channels 225 and 340, thus emulating a very anomalous peak regarding the expected shape of the peaks obtained for one emission from a silicon surface barrier detector. It can be seen how the function is able to reproduce the experimental distribution very well in spite of this very anomalous shape. This good agreement is clearly seen in the relative residuals shown in the figure, in which no significant structure appears.

In this case with such an anomalous shape it was necessary to use a number of knots larger than in the most common situations. Up to 11 knots were used to reproduce the overall shape of the "peak", distributed over a broad interval from the left-hand-most exponential region to close to the maximum. Trials using as few as 9 knots already gave very good results, with variations in area lower than 0.05% with respect to the area found in the fit shown in Fig. 6 (11 knots). With 9 knots, a χ^2_{red} of 1.123 was obtained as compared to the value of 0.980 found in the fit shown in the figure.

5. Conclusions

A new function is proposed for the analysis of alpha-particle spectra obtained from silicon detectors. The function is defined by parts such that at the edges the shape of the peak is preserved, in agreement with the expected response of this type of detector, while in the intermediate region a cubic splines-based term is introduced to form a smooth joint with the exponential and Gaussian regions (linear and quadratic in log-normal representations).

Detailed study of the function suggested that certain parameters put forwards in other proposals should not be considered. Probably, this was the reason for the important reduction in the number of knots needed to obtain good fits in the present work. This considerably improved the operativity of the program in its application to the analysis of alpha-particle spectra from semiconductor detectors. Moreover, the function is readily initialized, affording a very direct optimization process.

The function successfully reproduces extremely complicated experimental data distributions corresponding to spectra formed by peaks with shapes very different from those expected from silicon detectors. At same time, the program also produces very good results when it is applied to more conventional spectra, although in these cases the analysis process may be longer than those achieved with more usual functions.

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